### THE COLD CRUCIBLE VITRIFICATION PILOT PLANT: A KEY FACILITY FOR THE VITRIFICATION OF THE WASTE PRODUCED IN THE KOREAN NUCLEAR POWER PLANT

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#### ABSTRACT

Vitrification has been used for nuclear waste treatment for more than 35 years. It was initially applied to high level waste and the glass matrix is the international waste form for fission product solutions. Moreover, the successful operation of French vitrification facilities such as AVM in Marcoule or R7/T7 in La Hague has demonstrated with the vitrification of more than 4 Billions curies that it was also an industrial process.

The idea of using vitrification technologies to process low level and intermediate level waste was considered by SGN and KEPCO in 1995 and an optional study performed for KEPCO has demonstrated the cost-effectiveness of the solution applied to the waste produced by the Korean Nuclear Power Plant.

In 1997, after investigating high temperature technologies available on the market to process low and medium level waste produced by Nuclear Power Plants, NETEC (research institute of the Korean utility KEPCO) selected the cold crucible melter concept as the most promising vitrification technology for conditioning concentrates, ion exchange resins and combustible solids.

The Cold Crucible melter is an advanced vitrification melter developed by CEA for which electrical currents are directly induced in the melt from external high frequency inductors. The crucible is a simple, robust structure made of plain stainless steel, cooled by water and designed to be transparent to the electromagnetic field. The inherent advantages of the cold crucible (very long service life, low cost of the actual cold crucible and great flexibility in glass formulation) make it usable to treat both high level waste and low-level waste.

A joint NETEC-SGN-HDPIC development program was launched in 1997 to develop the industrial application of the CCM for the vitrification of the waste produced in the Korean Nuclear Power Plant.

The first step of the program was completed in 1998 and was dedicated to orientation tests in order to optimise the processing of the Korean waste in the CCM and to define the off-gas treatment system.

The second step of the joint collaboration, completed in October 1999, was devoted to the design and construction in Taejon of an industrial pilot plant This pilot has been operating since October 1999. The first vitrification campaigns are focused on the evaluation of the maximum throughput of the facility for the three types of waste and on the determination of the data necessary to establish process flowsheets for the waste produced in Nuclear Power Plants.

The main results obtained during these vitrification campaigns will be presented. The data related to the cost benefit associated with the use of this process for the treatment of the waste generated by the NPP will also be presented.

### INTRODUCTION

At the beginning of the nineties, difficulties in licensing waste disposal and subsequent impact on waste cycle cost lead many utilities to evaluate concepts of very limited Low Level Waste release. These concepts were based on volume reduction at the source and use of thermal treatment such as vitrification technology for the ultimate residue. If significant progress has been obtained by utilities on waste minimization at the source and if vitrification technology still appears one of the best candidate, it has not yet been implemented by any utility.

It will not be long now for such a demonstration, SGN (Cogema Group), NETEC (KEPCO) and HYUNDAI MOBIS (HYUNDAI group) are finalizing the development and industrialization program of LWR waste vitrification on the pilot scale 1 in Taejon, South Korea targeting final implementation of a vitrification facility on Ulchin site in 2004 / 2005.

This Korean project will benefit from the unique features of cold crucible melter technology developed since mid 80s in the French Commissariat à l'Energie Atomique Laboratories (CEA) and from the wide expertise acquired on commercial HLW vitrification in Cogema plants.

The challenge is to provide to Utilities a reliable innovative technology, which brings significant cost savings on waste management while complying with safety of workers and environment and allows optimization of the Waste Disposal Site life.

## WASTE FROM NUCLEAR POWER PLANTS

During their lifetime, light water reactors generate two categories of low and medium level radwaste, named as process and maintenance waste.

Process waste are mainly generated by treatment of water from reactor or ancillaries including spent fuel storage pools and some decontamination operation: usual effluent treatment technologies are based on distillation, ion-exchange, filtration or centrifugation. Typical process waste of PWR are organic bead resin ion-exchangers from primary and secondary circuits (blow-down resins), borated concentrates, sludge or filter cartridges. Typical waste of BWR are organic bead resins, concentrates and sludge containing different types of ion-exchange or filter media as organic powdered resins, diatomaceous earth, activated carbon, cellulose.

Maintenance waste are mainly solid dry active waste (DAW). They comprise damaged contaminated equipment which cannot be repaired or recycled and items such as contaminated clothes from operators, tools and plastic sheeting from maintenance work. A substantial part of the waste is burnable, most are compactable.

Process waste activity is concentrated in primary bead resins for PWR and some bead resins and sludge for BWR. Substantial part of resins (blow-down resins) and burnable DAW have a very low activity.

### NPP RADIOACTIVE WASTE MINIMIZATION PROGRAM

All over the world, utilities are making continuous effort to reduce LLW volume to be disposed of, acting at two levels, reducing waste production at source, and applying volume reduction treatment on generated waste.

The main driver of LLW minimization programs is the continous rising cost of disposal caused by the difficulties in public acceptance of new LLW disposal sites. Nevertheless, waste minimization program depends on some key issues:

### Waste Acceptance Criteria For Disposal

Waste acceptance criteria for disposal depend on the local conditions of the disposal site and the type of repository (surface or underground).

Volume reduction of very low active waste does not impact the disposal acceptance criteria because specific activity remains very far from upper threshold limits. For more active waste like some PWR primary resins or some BWR sludge blending with lower activity waste is advisable prior to disposal. (Special attention should be paid to long life beta emitters of activated products).

The final waste form acceptance criteria (leachability, mechanical strength...) for LLW are generally meet by cement, bitumen, polymers matrix or high integrity containers. For new matrix as glass, even though the final waste form is an improvement over current waste binders, some qualification work will be needed.

#### **Disposal Cost**

Very low disposal costs strongly dissuade volume reduction treatment of waste. Difficulties in site licensing lead to increased disposal costs. Disposal cost savings from volume reduction can finance the added cost necessary for waste treatment.

#### **Transport Regulation**

In theory there are three types of waste treatment facilities: on-site, mobile (site to site) or a centralized facility away from the NPP site. Prohibition of waste transport in some countries may preclude centralized treatment. If IAEA Transport recommendations are commonly in use in different countries, some complementary national rules may strongly impact waste management policy.

### **Cost Effectiveness of The Treatment Process**

When evaluating the cost effectiveness of the volume reduction treatment, two key criteria have to be assessed:

- the long term ability to maintain the throughput of the facility (reliability of the technology, maintainability of the equipment), taking into account a portion of the LLW waste has a significant activity
- the true waste volume reduction factor which must take into account the final waste package, the secondary waste generated by the process (sulfate and chloride salts) and

facility maintenance. The volume and the range of activity of the secondary waste are a key criteria to assess the cost effectiveness of the technology. Therefore, technologies relying on equipment with a short life (sensitivity to corrosion or wear) or having limited decontamination factors may impact dramatically the waste cost by generating associated waste treatment and disposal costs.

If some industrial thermal treatment technologies have been demonstrated for volume reduction of lower activity waste such as Dry Active Waste or blow-down resins of PWR (in dilution with DAW). Proven industrial solutions for higher activity waste are still pending.

Besides the quality of containment, public acceptance for vitrification is high, although pure incineration technologies suffer a more controversial image

## COLD CRUCIBLE MELTER VITRIFICATION TECHNOLOGY

Vitrification is recognized worldwide as the best available technology for high level waste processing.

CEA scientists and Cogema operators have gained more than 30 years of expertise in HLW vitrification. These efforts have resulted in a unique, high-performance in-line vitrification process, which is now clearly one of the best solutions in terms of industrial maturity, efficiency and cost.

Three vitrification plants are currently in commercial operation in France for HLW immobilization: AVM (1978), R7 (1989) and T7 (1992) in France (UK: WVP, 1992). All are based on the same two-step process, which consists of calcination of an aqueous solution prior to melting with vitreous additives in a metallic crucible heated by induction.

More than 10,000 canisters and 4000 t of HLW glass representing several billion Ci, have been produced to date. The waste form has been licensed by the French Regulatory Body and international Safety Authorities from UK, Belgium, Japan, Germany, Switzerland and the Netherlands.

The basic principles of the process are:

- two step process: calcination and glass melting,
- metallic melter heated by induction which means:
  - light and compact melter,
  - melter independent from induction heating system,
  - melter easy to replace or evacuate (in same size glass-canister type)

This process is quite satisfactory for concentrated HLW solutions of modern reprocessing plant but, its throughput is limited and it is not appropriate for waste, which are corrosive or require temperatures higher than 1100°C (e.g. refractory glass).

CEA has therefore been developing the cold crucible melter technology since the mid-80s in order to expand the range of glass and waste that can be vitrified while retaining the main successful features of the AVM/AVH process, namely the metallic melter and induction heating. In the cold crucible concept, electrical currents are directly induced into the glass melt from external high frequency inductors. As the molten glass is directly heated, the

crucible wall can be cooled. The cold structure causes a thin layer of solidified glass material to coat the surface in contact with glass, thereby protecting it from the corrosive melt. Since the energy source is outside the melter, the recurring problem of electrode corrosion is avoided. In addition to providing a considerable lifetime for the melter, this technology overcomes the temperature limits imposed in more conventional systems with Inconel electrodes (1050 to 1150 °C) and accommodates corrosive glass or refractory melt (up to more than 2000° C) that could not otherwise be processed. The glass is poured periodically through a cooled bottom drain valve, which, coupled with efficient melt agitation, makes the technology well adapted to the removal of all kinds of insoluble. The crucible itself is a simple, robust structure made of plain stainless steel sectors, cooled by water. The structure in sectors helps to develop an electromagnetic field in the crucible.

The main features of this process include all the benefits from direct induction heating of molten glass in a small metallic cold wall crucible, i.e.: increased melter life time (no sensitivity to sulfate corrosion), flexibility of glass elaboration (due to a wide range of acceptable temperature), high density of power, small lay-out and very low generation of secondary waste.

If the cold crucible is in the continuity of the compact metallic melter, a significant advantage is that its high density of power does not require a calcination step prior to melting. Evaporation, calcination and vitrification of solutions can be performed in the same equipment.

## APPLICATION OUTSIDE THE NUCLEAR INDUSTRY

Specific features of the cold crucible technology look promising for some applications outside the nuclear industry. Since 1995 a cold crucible with a diameter of 600 mm has been operating successfully in the glass industry with a throughput of 50 kg/h. A new cold crucible melter with diameter of 1200 mm was installed in 1998 with a glass throughput of 250 kg/h to 400 kg/h.

## **APPLICATIONS FOR H IGH LEVEL WASTE**

#### **CORA Project**

In 1997, ENEA, the Italian Atomic Energy Agency, awarded a contract to SGN for the design and construction of a facility to vitrify the legacy HLW waste stored in the Salluggia Center. The waste was generated by Candu and MTR spent fuel reprocessing and represents a total volume of about 200 m<sup>3</sup> with some Hg traces and significant quantities of aluminum. The melter will be fed with an aqueous solution. The challenge was to implement the process in existing cells and particularly to generate less than 100 canisters to avoid new storage building construction. Plant should be commissioned in 2004.

#### **U-Mo Project**

COGEMA plans to apply the cold crucible technology in the La Hague plant for the vitrification of high level liquid waste coming from reprocessed spent Mo-Sn-Al fuel. This particular waste stream has been selected for vitrification in the cold crucible because of its high Molybdenum content which makes it very corrosive and which requires a specifically tailored glass formulation in order to obtain high waste loading factors. This glass formulation

requires high operating temperatures (approximately 1300°C), which cannot be reached with the standard hot induction melters currently operated in the La Hague vitrification facilities. The unlimited melter lifetime, the fact that there is virtually no upper bound on the operating temperature and the fact that the melt is mechanically stirred will practically suppress the operating constraints that would be encountered with standard technologies for this type of hard-to-process waste. The process and the different technologies associated (pour valve, process instrumentation,...) are currently being qualified on a full-scale prototype at the CEA pilot facility in Marcoule. The glass formula is also being qualified in order to meet standard waste acceptance criteria for final disposal. Hot operations are scheduled to start in the R7 facility in 2004 [1]

# APPLICATIONS FOR LOW AND MEDIUM LEVEL WASTE

### The Korean Project

The subsurface disposal concept (more flexible to waste concentration) and the difficulties of site selection led KEPCO to evaluate various efficient thermal treatment technologies for NPP waste and in particular the one ensuring vitrification of ion-exchange resins, concentrates and DAW.

In 1995 and 1996 studies from KEPRI and SGN were performed to assess different melter technologies, examine how innovative high temperature technologies could be implemented to achieve a good volume reduction of the waste, evaluate and compare these technologies from the technical and economical viewpoints.

Technical and economical assessment conclude that the best candidate was the cold crucible for all the waste except for spent cartridges filters and non combustible waste which could be treated by plasma torch.

KEPRI (now NETEC) conclude its investigation on high temperature technologies available on the market by selecting the cold crucible melter concept as the most promising for conditioning concentrates, ion exchange resins and combustible solids.

A joint KEPRI-SGN development program started in 1997 with the active support of CEA and COGEMA to develop cold crucible incineration/vitrification of NPP waste on an industrial scale through a multi-step program.

#### Phase 1:

Phase 1 of the program, which was completed in 1997, was dedicated to orientation tests on laboratory scale equipment and on a CEA pilot facility with a 300 mm diameter CCM. The main target of this phase was to evaluate the feasibility of direct incineration/vitrific ation in the cold crucible and to finalize the design of the future pilot to be erected in Korea. Main investigations were aimed at optimizing the combustion process and defining the optimum off-gas treatment system.

The tests performed with Ion Exchange Resins provided valuable information on the IER processing capability of the CCM and on the operating conditions. The process atmosphere must be oxidizing to avoid glass reduction by carbon from the resins and combustible waste. An oxygen excess of 25 % was found to be sufficient to avoid such phenomena. The glass

surface temperature was limited to 1200°C to limit the radionuclide volatility during the combustion. In these conditions, the throughput of the 300 mm- CCM is 12 kg/h with IER.

The dust content at the outlet of the CCM ranged from 1 to 10 g/Nm3 depending on the type of resin, the water content and the type of vitrification additives. A high Temperature Filter equipped with ceramic candles was tested to remove the dust from the off-gas. Satisfactory results were obtained, and the filter efficiency was between 99.6 and 99.8 % at 200°C. The high temperature filter decontamination factors were found to be 2000 for Co and 4000 for Cs.

Main process performances and key process parameters have been assessed in this step for ion-exchange resins mainly, nevertheless the optimization of the glass matrix and combustion of dry active waste was considered to be less critical and to be performed in phase 3 on the Taejon Pilot facility.

### Phase 2:

Phase 2 involved the design and construction of the facility in Taejon, South Korea and ended successfully in October 1999, with the inauguration of the facility.

This facility is presented on figure 1 and includes:

- a waste preparation and feeding unit including preparation units (dryer and shredding) storage hoppers and transfer and metering system
- a 550 mm diameter CCM equipped with a bottom pouring valve, oxygen injectors and waste feeding system
- an off gas-treatment system including a pipe cooler, a high temperature filter, a postcombustion center, a packed bed scrubber, re-heater, HEPA filter and a De-NOX unit.

Four types of surrogate waste can be processed in the facility:

- Ion Exchange Resins
- Dry Active Waste (e.g. plastic and cellulose waste)
- Borated concentrate
- Sludge

#### Phase 3:

Phase 3 began with the start-up of pilot facility operation in October 1999. The development program performed by NETEC in this phase with support of SGN / Cogema / CEA and HYUNDAI MOBIS simulates the behavior of a commercial vitrification plant as closely as possible and intends to finalize the design parameters and design options.

During the first campaign performed between November 99 and May 2000, about 1,500 kg of IER have been processed at a throughput varying between 20 and 40 kg/h. The dust generated at the outlet of the CCM was observed to be between 2 and 20 g/Nm3 and was efficiently captured by the High Temperature filter. The CS capturing efficiency of this filter was found to be 99.9% during the tests. All regulated gases at the stack inlet were well below the environmental regulation limits.

Analysis performed on the glass have revealed a homogeneous composition. US EPA TCLP tests were performed on glass samples obtained during 5 different resins tests and all the leachates from the samples are well below the regulatory limits, i.e. Universal Treatment Standards for 14 metals (As, Ag, Ba, Be, Cd, Cr, Hg, Ni, Pb, Sb, Se, Th, V and Zn) from the Federal Register of 40CFR268.

These initial results have confirmed the Marcoule phase 1 campaign observations and in particular the throughput escalation from 12 kg/h (in a 300 mm diameter CCM) to 40 kg/h of ion-exchange resins (in a 550 mm diameter CCM).

## CONCLUSION

Significant progress in vitrification of NPP waste has been made through the collaboration between NETEC/KEPCO, SGN/Cogema/CEA and HYUNDAI MOBIS. The first campaign performed on the Taejon pilot plant has demonstrated the functionality, reliability, high productivity and ability to maintain differential pressure during the operation.

The implementation of the versatile Cold Crucible Melter Technology in an industrial facility will soon be a reality and utilities will benefit from the important advantages associated with the vitrification of the waste produced by Nuclear Power Plants.

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Fig. 1: Flow sheet of the Taejon Vitrification Facility